ESTIMATION OF THE BIOACCUMULATION OF MERCURY BY BLUEGILL SUNFISH IN EAST FORK POPLAR CREEK

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ESTIMATION OF THE BIOACCUMULATION OF MERCURY BY BLUEGILL SUNFISH IN EAST FORK POPLAR CREEK

INTRODUCTION

Studies conducted by Van Winkle, et al., (1984), on East Fork Poplar Creek (EFPC) documented the system to be contaminated by mercury originating from the Department of Energy's Y-12 plant. Specifically, the study showed (1) that the fine sediments in the stream were significantly contaminated with mercury; the mercury contamination was highest near the Y-12 discharge and decreased in downstream sediments, and (2) fish in the system had mercury concentrations in muscle tissue exceeding FDA limits of 1.0 ppm.

As a result of this study and other water quality concerns (i.e., PCB contamination), the Oak Ridge Task Force, a work group consisting of representatives from the State of Tennessee, EPA, TVA, ORNL, DOE, and other non-regulatory groups, was formed to evaluate the problem and make recommendations for remedial action. Several studies were initiated under the direction of the group through funding from DOE (TVA, 1985a, b, c, d, e) to assess the situation. Additional studies were proposed by ORNL to further define contamination problems.

One of the proposed studies, "Role of Current Discharges from Y-12 Plant in Regulating Mercury and PCB Uptake by Fish in East Fork Poplar Creek" (Elwood and Turner, 1985), was initiated in July 1986. The Oak Ridge Task Force requested the Tennessee Valley Authority to act as

an independent reviewer of the project data; to use the project data to calibrate a simple steady-state mercury bioaccumulation model and provide a final technical report on the results. This request resulted in an interagency agreement between DOE and TVA, (contract number DE-A105-860R21596).

DESCRIPTION OF STUDY AREA

East Fork Poplar Creek

The source of EFPC originates within the Y-12 plant area at Oak Ridge National Laboratory, near Oak Ridge, Tennessee (Lat 35° 57' 58", Long 84° 21' 30"). During the study period (i.e., July through December 1986), the flow in EFPC averaged 26 cfs and the minimum daily discharge was 17 cfs for several days in July and August (USGS water year 1986 to 1987). The USGS records note that the Y-12 plant may contribute up to 20 cfs and the west end sewage treatment plant of the City of Oak Ridge may add up to 10 cfs. Figure 1 shows a generalized map of the area including the location of the six study sites selected for fish uptake studies (see Elwood and Turner, 1985). Site 5 is located just below the sewage treatment plant. Sites 1, 2, 3, and 4 were in a reach of EFPC dominated by flow from the Y-12 plant.

Mercury in Sediment

Mercury adsorption to suspended particles and sediments occurs by adsorption-desorption equilibria. The sediments of EFPC contain high concentrations of total mercury which has been shown to be associated with sediment fractions < 0.125 mm (Van Winkle, et al., 1984). Subsequent work performed by TVA under contract to DOE showed 10 percent of mercury in the sediment associated with the > 0.5 mm fraction, 50 percent with the .062 mm - 0.5 mm fraction, and 40 percent with the < .062 mm fraction (TVA, 1985b).

Table 1 provides a summary of mercury concentrations and other constituents in sediment for selected sampling stations within EFPC. Inspection of table 1 shows that in the upper reaches of EFPC, the sediment has more mercury in the < 0.125 mm fraction and that the concentrations of mercury decrease with stations proceeding downstream. While analysis of mercury in silt/clay fractions is a good technique to evaluate mercury transport, it should not be confused as being representative of total mercury contamination in the system. These results would support the assumption that total pounds of mercury in the system is likely to be higher near the upstream source and to decrease in downstream reaches. The concentrations in downstream areas (table 1) are due to transport of scoured clay and silt materials from upper reaches of EFPC. Figure 2 shows that relationship between mean stream velocity and sediment transport as a function of particle size.

THEORETICAL CONCEPTS AND TECHNICAL APPROACH FOR MODELING BIOACCUMULATION OF MERCURY BY FISH

Mercury Uptake by Aquatic Organisms

The principal pathways for uptake of mercury by fish are from food and water. The few controlled experiments that are reported in the

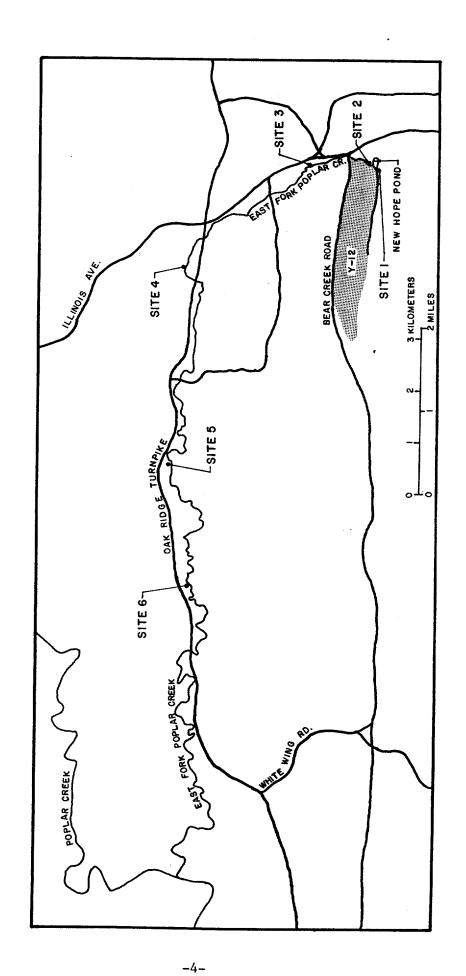


FIGURE 1 LOCATION OF SAMPLING SITES ON EFPC (Modified from Van Winkle, 1984)

TABLE 1

TOTAL Hg AND OTHER CONSTITUENTS
IN EFPC SEDIMENTS COLLECTED JULY 1986

	<2 1	mm Size F	raction			<0.125 mm	Size Frac	tion
Site	Carbon (%)	Sulfur (%)	Water (%)	Hg (ug/g)	Hg (ug/g)		PCB-1260	Σ PCB
1	13.9	0.28	58	128	125	1.6	2.3	3.9
NHP Sfc*	12.0	0.34	61	146	128	2.1	2.8	4.9
2	7.79	0.19	65	63	62	0.51	0.94	1.45
3	3.75	0.15	65	38	34	0.19	0.67	0.86
4	3.52	0.096	64	58	45	0.38	0.71	1.09
5	2.40	0.064	62	39	35	0.24	0.49	0.73
6	1.23	0.046	50	28	18	0.13	0.26	0.39

^{*} New Hope Pond surface sediments (top 1 cm)

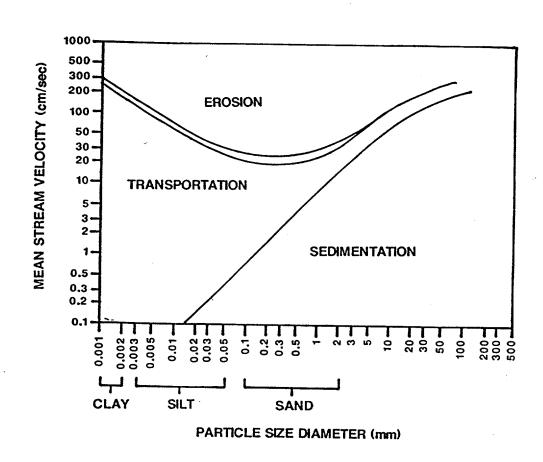


FIGURE 2 RELATIONSHIP BETWEEN STREAM VELOCITY,
PARTICLE SIZE, AND THE REGIMES OF
SEDIMENT, EROSION, TRANSPORT, AND
DEPOSITION. (FROM: GRAF, 1971)

literature provide conflicting conclusions regarding the importance of one mechanism over the other. Also, the extrapolation of laboratory results to the natural environment may not be accurate because the exposure levels employed in the lab often greatly exceed those found in nature.

Figure 3 shows idealized relationships among the principal pathways and processes affecting mercury uptake by fish. Note that the water source is composed of two vectors, i.e., soluble mercury and mercury sorbed to suspended sediment. Direct uptake from contaminated suspended sediment is a mechanism that has not been previously described in the literature. The figure also shows several intrinsic and extrinsic processes which control accumulation of mercury by fish.

Several investigators maintain that the direct uptake of contaminants by fish is proportional to metabolic rate (e.g., Boddington, et al., 1979; Neely, 1979, McKim and Heath, 1983; McKim and Goeden, 1982). The uptake rate of pollutants by fish should fall within limits set by those factors that control metabolism and growth, as modified by environmental factors, such as temperature and food availability (Norstrom, et al., 1976). Eberhardt (1975) recommended that the log of residue concentration be expressed as a function of log body weight to reduce variability among samples. Such correlations in field data suggest that the mechanism of residue uptake is ultimately linked to the metabolic activities of the fish.

Theoretically, if uptake is directly proportional to metabolic rate, the concentration factors for food should not exceed measures of

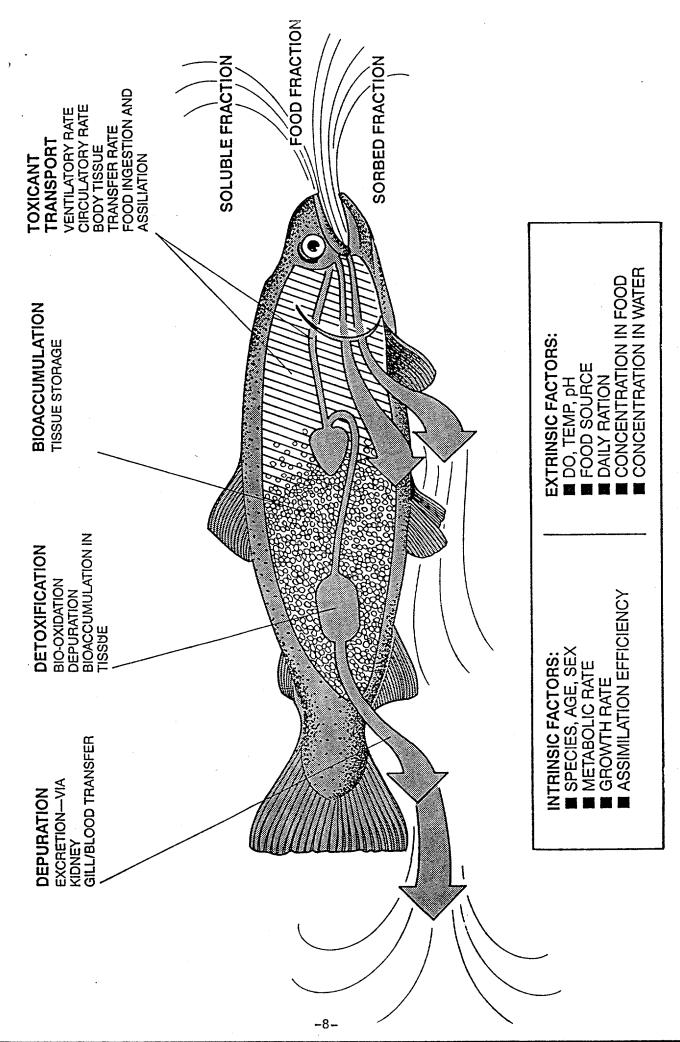


FIGURE 3 PATHWAYS AND PROCESSES INVOLVED IN UPTAKE BY FISH

gross growth efficiency (Slobodkin, 1970). The growth efficiency of fish is about 80 percent. Hamelink and Spacie (1977) state that any dietary uptake efficiency greater than 80 percent will lead to the accumulation of residues. For pesticides, the literature indicates that uptake tends to occur over the entire body surface of smaller organisms, but may be more confined to particular sites of organisms with more substantial integuments (body surfaces), such as fish (Ferguson, et al., 1966). like manner, residues of mercury can enter fish directly from the water through the gill surfaces, as well as with ingested food. Entry through the integument may be more difficult. The relative importance of these processes of direct uptake from water, assimilation from stomach contents, and diffusive loss to water is not clearly understood. The relative importance of each process would vary with environmental conditions and with the nature of the compound and the organism (Kerr and Vass, 1973). Determining factors as to which of the processes are most important are:

- 1. The persistence, distribution, and concentration of the chemical in the water as contrasted to that in the food;
- 2. The stability of the chemical in the digestive system compared to that in the water;
- The distribution coefficients of the chemical between the gill surface and blood versus the digestive system and blood of the organisms; and,
- 4. The relationship to metabolism of the different routes of a chemical (Kenaga, 1975).

The bioavailability of a chemical affects both initial uptake rate and steady-state concentration in fish tissue.

Depuration of Mercury by Aquatic Organisms

Depuration is the process of elimination of a material from an aquatic organism. Depuration rates define this elimination process as a function of time. Likewise, depuration can be expressed as half-life or the time required to eliminate half of the material from the organism.

Half-life values for loss of mercury vary greatly in the literature. For fish, they range from about 30 days to 180 days (Burrows, 1973).

The half-life of mercury may be dependent upon metabolic functions, such as availability of mercury to the blood stream, differences in the types of biochemical bonding, and the anatomical locations of mercury-bearing tissues. Also, differences in rates of depuration from fish contaminated through their food or directly from the water, have been reported (Jarvinen, et al., 1976). Seasonal changes may also occur. For example, temperature apparently does not influence depuration rate, although it does influence the uptake rate. Whether this mechanism holds true for protein bonding compounds such as mercury is questionable.

Whether the partition hypothesis can be applied to depuration is questionable. The hypothesis requires that, given clean water, mercury residues will eventually disappear completely from the organism. This is not supported by most experimental evidence for higher organisms (Young, et al., 1971). Most data suggest that clearance of a residue

from an organism depends not so much on the possibility of a tissue-water distribution equilibrium, but on the time taken to attain it (Sprague, et al., 1971; Addison, 1976).

To understand depuration, it is important to know the metabolic fate of the chemical tested. A principal mechanism occurring within the organism is the conversion of the parent compound to more water soluble metabolites. For example, in the case of DDT, this conversion is DDT to DDD and DDE. However, the elimination of mercury compounds is less clearly understood. Burrows (1973) followed the depuration of mercury by bluegill for a period of 100 days. Inspection of Burrows' data shows two distinct depuration rates: one fast (half-life ~ 35 days) followed by a slower rate (half life ~ 180 days). Burrows' work also demonstrate no significant difference in depuration rates of fed vs. starved fish. Hartung (1976) suggests that the initial rapid loss rate is a consequence of the method of dosing (i.e., short term/high concentration) and that fish chronically exposed do not demonstrate bi-phasic depuration rates.

METHODS

Mathematical Models

There are two basic modeling approaches for predicting the uptake of xenobiotic chemicals by fish:

 Those that use the equilibrium approach (partition coefficients; bioconcentration factors, and other empirical relationships), and Kinetic models (models that employ uptake and depuration rate constants from water and food and that may also include growth and other bioenergetic factors).

Each approach may be useful, but, in general, kinetic models provide a more versatile tool for evaluating and understanding dynamic situations.

A compartmental model can demonstrate the pathways of mercury exchange among the biotic and abiotic components of the aquatic environment and the kinetic and equilibrium constants that control the distribution of mercury between compartments. For example, consider a finite column of water that receives and releases water at a flow rate of Q and covers a uniform deposit of mercury-contaminated sediment. The mercury concentration in the water compartment can be described by the following mass balance equation:

$$\frac{dC_{w}}{dt} = \frac{C_{in} - C_{w}}{t_{o}} + \frac{J_{s}}{Z}$$
 (1)

Where:

 C_W = mercury concentration in water in the container (mg/1)

 C_{in} = mercury concentration in water in the influent (mg/1)

t = time (days)

 t_0 = hydraulic retention time (days)

J_s = mass of mercury being released from the sediment per unit area and time (mg/sq cm-day)

Z = depth of water column (cm)

The <u>mass balance</u> equation for mercury concentration in a fish confined to the water compartment follows:

$$\frac{dC}{dt} = k_w C_w + AERC_f - k_d C \qquad (2)$$

Where:

C = mercury concentration in fish (ug/g)

 k_w = fish uptake rate constant through mercury-contaminated water (day⁻¹)

AE = assimilation efficiency for food (percent)

R = daily food ration (g/g day)

 C_f = residue concentration in food (ug/g)

 $k_d = depuration rate (day^{-1})$

If $\mathbf{C}_{\mathbf{w}}$ and $\mathbf{C}_{\mathbf{f}}$ are assumed to be constant, Equation 2 can be integrated to yield:

$$C = \frac{k_{\text{w}}C_{\text{w}} + AERC_{\text{f}}}{k_{\text{d}}} (1 - e^{(-k_{\text{d}}t)}) + C_{\text{o}} (1 - e^{(-k_{\text{d}}t)})$$
(3)

Where:

 C_{0} is the mercury concentration in fish at t = 0.

(Note: If C_W is not constant, Equation 1 must be integrated first and incorporated into Equation 2. The same is true for C_f except that another mass balance equation would be used for C_f .)

The biological half-life of mercury t(1/2) in fish can be determined by:

$$t_{1/2} = -\ln 0.5/k_d$$
 (4)

To account for the effect of the size of fish, Wilson (1969) used a correlation between the rate of uptake or depuration and the weight of fish. He replaced the rate terms, such as $k_{\rm w}$ and $k_{\rm d}$, in the equation similar to Equation 3 with his correlation relationships. Another

approach to account for the size of fish is the use of the growth rate concept which can be incorporated into the model in a similar fashion (i.e., $k' = k_d + g$).

The approach discussed here is not presented or proposed to be exact or inclusive of all variables that affect mercury concentrations in fish. The purpose is to show that the relative importance of food and water as sources of uptake can be quantified from the magnitudes of the numerical coefficients preceding \mathbf{C}_f and \mathbf{C}_w . In cases where uptake from the water is shown to be a significant source and \mathbf{C}_w varies significantly with time, these equations may have to be coupled with hydraulic models specific to EFPC in order to predict mercury accumulation in fish. Evaluation of more detailed kinetic models, i.e., TOXIWASP, can provide further refinement of information needs and predictions of how environmental parameters affect mercury accumulation in fish.

Kinetic Model Estimation of Food and Water-Derived Mercury Residues in Fish

The development of a model which could be used to estimate and quantify the kinetics of mercury accumulation and release in fish was pursued assuming steady-state conditions.

Initial Steady-State Model Calibration

To evaluate mercury uptake solely from water sources, the food uptake component of Equation 2 is omitted. The model shown in Figure 4

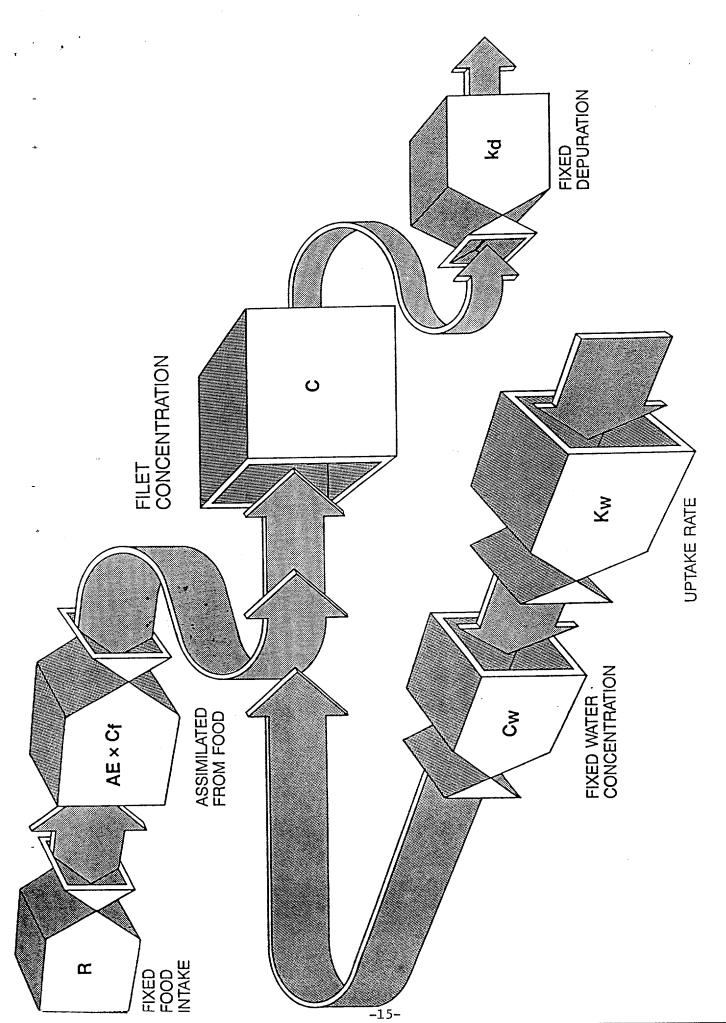


FIGURE 4 ONE COMPARTMENT BIOACCUMULATION MODEL

assumes first order kinetics of the form:

$$dC/dt = k_w * C_w - k_d * C$$
 (5)

If dc/dt is steady-state, then:

$$\emptyset = k_w * C_w - k_d * C_{ss}$$

Where:

 $C_{w} = total water mercury concentration (ug/ml)$

C = mercury concentration in bluegil1 filet (ug/g)

 $k_{\rm u}$ = uptake rate constant day⁻¹)

 $k_d = depuration rate constant (day^{-1})$

 C_{cc} = steady state concentration (ug/g)

t = time (days)

RESULTS AND DISCUSSION

The model above was calibrated using the data collected by ORNL from in situ caged fish and studies conducted at EFPC. These studies were conducted using hybrid bluegill/green sunfish at five locations within the system. Table 2 provides a summary of total and soluble mercury data in water at each site by sampling date. Mercury concentrations in filets were monitored periodically over a period of 50 to 110 days. These data are presented in Appendix A. The limitations of these data sets include:

 The uptake experiments were not conducted for a sufficient period of time to demonstrate that a steady-state concentration of mercury was reached at all locations.

SUMMARY OF TOTAL AND SOLUBLE MERCURY IN EFPC WATER AT CAGED FISH STUDY SITES (UG/L HG)

Sol Tot Sol Tot Tot 0.2 1.2 0.71 1.10 0.06 0.70 0.11 0.6 0.11 1.0 0.12 0.56 0.06 0.68 0.15 0.56 0.11 0.68 0.11 0.49 0.08 0.43 0.16 0.63 0.09 0.51 0.51 0.53 0.09 0.94 0.05 0.09 0.94 0.05 0.13 0.71 0.74 0.05	Site 1	1 1	V)	Site	2	Site 3	က	Site 4	4	Site	₂	Site 6	9
0.2 1.2 0.7¹ 1.10 0.06 0.70 0.1 1.0 0.12 0.56 0.06 0.68 0.15 0.05 0.11 0.68 0.11 0.49 0.08 0.43 0.16 0.63 0.09 0.7 0.51 0.09 0.94 0.05 0.51 0.09 0.94 0.05 0.29 0.09 0.94 0.05 0.29 0.09 0.09 0.09 0.09 0.13 0.71 0.74 0.05 0.59	Tot Sol Tot Sol	1	ot Sol	Sol	•	Tot	Sol	Tot	Sol	Tot	Sol	Tot	Sol
0.11 0.6 0.11 1.0 0.12 0.56 0.06 0.68 0.115 0.56 0.111 0.53 0.11 0.49 0.08 0.43 0.16 0.63 0.09 0.51 0.51 0.09 0.094 0.05 0.37 0.04 0.29 0.13 0.71 0.082 0.74 0.05 0.59	2.6 1.3 1.4 0.3		4 0.3	0.3		1.4	0.2	1.2	0.71	1.10	90.0	0.70	
0.12 0.56 0.06 0.68 0.15 0.56 0.11 0.53 0.11 0.49 0.08 0.43 0.16 0.63 0.09 0.51 0.51 0.09 0.094 0.05 0.37 0.04 0.29 0.13 0.71 0.082 0.74 0.05 0.59	3.7 0.9 1.8 0.6	1.8	9.0 8	9.0		0.61	0.1	9.0	0.1	1	i	1.0	0.0
0.15 0.56 0.11 0.53 0.11 0.49 0.08 0.43 0.16 0.63 0.09 0.51 0.51 0.09 0.94 0.05 0.37 0.04 0.29 0.13 0.71 0.082 0.74 0.05 0.13 0.71 0.082 0.74 0.05	2.76 0.58 1.30 0.37		30 0.37	0.37		0.81	0.12	0.56	90.0	1	1	0.68	0.02
0.11 0.49 0.08 0.43 0.16 0.63 0.09 0.51 0.51 0.09 0.094 0.05 0.37 0.04 0.29 0.13 0.71 0.082 0.74 0.05	7.5 6.1 1.30 0.31		30 0.31	0.31		0.80	0.15	0.56	0.11	1	i	0.53	0.14
0.16 0.63 0.09 0.51 0.09 0.94 0.05 0.37 0.04 0.29 0.13 0.71 0.082 0.74 0.05 0.59	2.8 0.18 1.4 0.19		4 0.19	0.19		1.5	0.11	0.49	0.08	1	1	0.43	0.06
0.09 0.94 0.05 0.37 0.04 0.29 0.13 0.71 0.082 0.74 0.05 0.59	2.82 0.59 1.25 0.28	1.25 0.2	0.2	0.28		0.99	0.16	0.63	60.0	1	:	0.51	0.0
0.09 0.094 0.05 0.37 0.04 0.29 0.13 0.71 0.082 0.74 0.05 0.59	1.80 0.26 1.30 0.09	1.30 0.0	30 0.09	0.09		:		i i	:		;	1	ı
0.09 0.94 0.05 0.37 0.04 0.29 0.13 0.71 0.082 0.74 0.05 0.59	5.00 0.80		•	:		:	:	:	! !	:	!	•	•
0.37 0.04 0.29 0.13 0.71 0.082 0.74 0.05 0.59	:	:	!	1		0.93	60.0	0.94	0.05	;	<u>.</u>	:	•
0.13 0.71 0.082 0.74 0.05 0.59	:	:	:	;		1	;	\$ 1	!	0.37	0.04	0.29	0.
0.13 0.71 0.082 0.74 0.05 0.59	3.62 0.45	3.62 0.4				;	:	:	:	;	;	;	•
	3.62 0.75 1.67 0.32	1.67 0.3			,	1.00	0.13	0.71	0.082	0.74	0.05	0.59	0.0

16

 $^{\mathrm{1}}\mathrm{Not}$ used in average for model

- 2. No depuration (loss rate) studies were included.
- Water column methylmercury data were insufficient to establish exposure concentrations at all stations.

Soluble mercury data was chosen for model calibration due to the limited amount of methylmercury data for the study sites and the fact that the data showed a strong relationship between average soluble mercury concentration in the water column at each site and the total mercury concentration in filets. Although most previous research suggest that methylmercury is the main source of mercury bioconcentration by fish, inorganic mercury may also be accumulated. Specifically, the soluble mercury data for site 2 was used to derive model coefficients and calibrate the model.

Figure 5 shows a plot of the mean and upper and lower 95 percent confidence limits for site 2 data. There was no significant difference in the mean values for the last three data points, and the fish were assumed to have reached steady state concentration.

Estimation of the Depuration Rate Constant k

The depuration rate constant k_d can be estimated from a plot of the concentration in the fish at time t divided by the steady state concentration vs. time. Figure 6 shows such a plot for Site 2 data. The value k_d was estimated from the relationship:

$$k_d = -\ln 0.5/t_{50}$$

Where:

t₅₀ = the time to reach half saturation (i.e., 50 percent of Css).

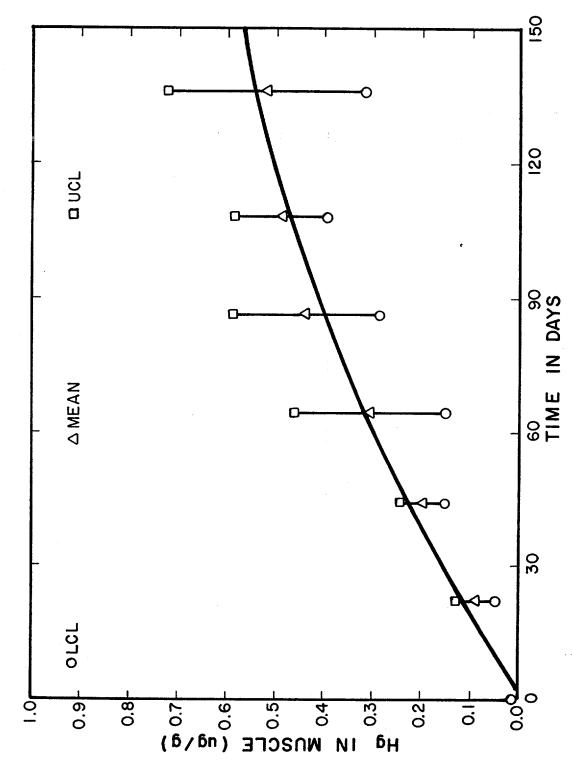
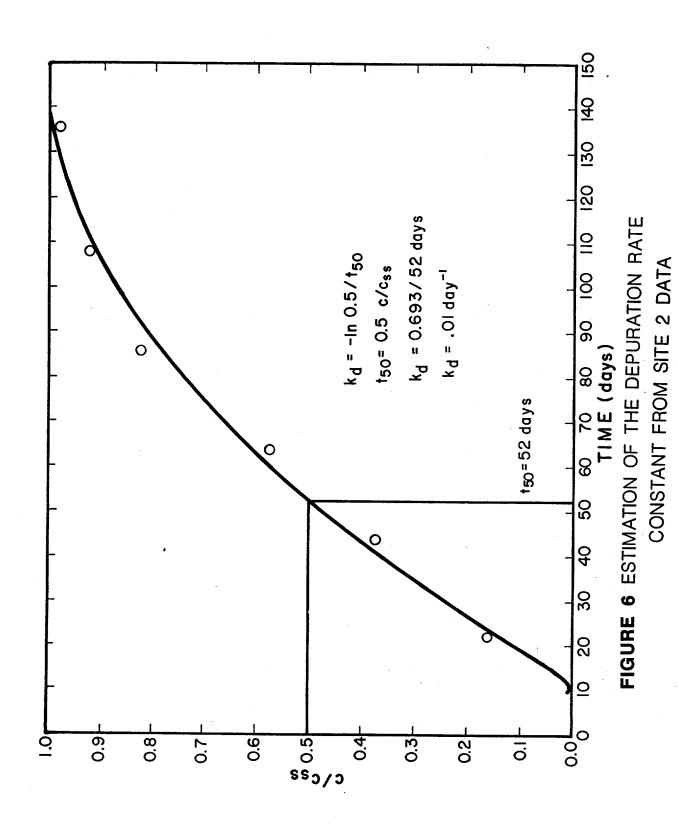


FIGURE 5 MEAN AND 95% CONFIDENCE INTERVALS FOR MERCURY IN BLUEGILL FILET VS TIME FOR SITE 2 DATA



The estimated k_d value of 0.01 d^{-1} is approximately one order of magnitude larger than k_d values reported in the literature for other species of fish, however, accepting steady-state conditions as demonstrated by the data, this value was used to calibrate the model.

Estimation of the Uptake Rate Constant k

The uptake of mercury by fish during continuous exposure is not easily predicted because depuration begins to occur as soon as the uptake begins, so that uptake and elimination interact to determine the storage of mercury at any one time. One method used to estimate k_w is to manually measure the tangent to the uptake curve ($\Delta C/\Delta t$) at several points. Each tangent is an approximation of dC/dt that can be used to calculate a value of k_w using the method of Blanchard et al., 1977. Table 3 shows the results of these calculations for site 2 data. The average k_w value was estimated to be 22.16 d⁻¹.

Model Calibration

Employing the value for k_d and k_w estimated from site 2 data and the average water column soluble mercury concentration for site 2, the model (equation 5) was calibrated. Table 4 shows the observed versus predicted mercury in filets for site 2 and sites 3, 4, and 6 (sites 1 and 5 data were insufficient to model). Figure 7 shows a correlation plot of the data from table 4. The correlation between observed and predicted values is strong (r2 > 0.9) and the variance is well within the range of the 95 percent confidence limits for the data collected. Because the

TABLE 3 $\begin{tabular}{lll} \hline ESTIMATION OF $k_{\pmb{w}}$ FROM SITE 2 \\ \hline SOLUBLE MERCURY DATA 1 \\ \hline \end{tabular}$

Exposure Time (Days)	Tangent $(\Delta C/\Delta t)$	^k d-1)	C (ug/g)	cw ² (ug/ml)	Estimated k _W
20	.00375	.01	.075	.00032	14.06
40	.0050	.01	.175	.00032	21.09
80	.0056	.01	.400	.00032	30.06
120	.0025	.01	.500	.00032	23.43
Mean					22.16

The tangent to the uptake curve was determined grapahically at each observation time. The rate constant k_d was derived as previously described. $k_W = (\Delta c/\Delta t + k_d C) CW$

² Average soluble mercury data for Site 2 (N = 7)

TABLE 4

OBSERVED VS PREDICTED¹ MERCURY IN MUSCLE TISSUE (ug/g)
BASED ON MEAN SOLUBLE MERCURY AT SELECTED SITES

Site 2 (0.32 ug/l Hg)	Site 3 (0.13 ug/l Hg)	Site 4 (0.082 ug/l Hg)	Site 6 (0.06 ug/l Hg)
Obs. Pred.	Obs. Pred.	Obs. Pred.	Obs. Pred.
.087 .140	.037 .057	.027 .036	.023 .026
.184 ² .140	,-		
.198 .252	.075 .103	.039 .065	.034 .0473
.305 .335	.114 .136	.078 .086	.044 .063
.376 ² .367	.348 ² .149	.254 ² .0941	.127 ² .069
.480 .409	.165 ² .166	.132 .105	.046 .077
.488 .468			.056 .088
.518 .527			
	(0.32 ug/1 Hg) Obs. Pred. .087 .140 .184 ² .140 .198 .252 .305 .335 .376 ² .367 .480 .409 .488 .468	(0.32 ug/l Hg) (0.13 ug/l Hg) Obs. Pred. Obs. Pred. .087 .140 .037 .057 .184 ² .140 .198 .252 .075 .103 .305 .335 .114 .136 .376 ² .367 .348 ² .149 .480 .409 .165 ² .166 .488 .468	$\begin{array}{cccccccccccccccccccccccccccccccccccc$

 $^{^{1}}$ Model calibrated from Site 2 data - assumes no food uptake. $\rm K_W = 22.16~d^{-1},~K_d = .01~d^{-1}$

 $^{^{2}}$ Data from fish collected outside cage

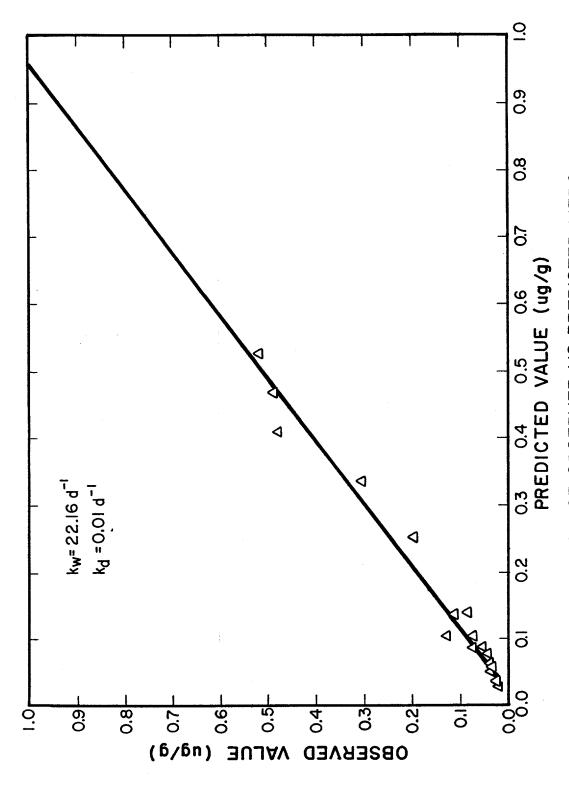


FIGURE 7 CORRELATION OF OBSERVED VS PREDICTED MERCURY TISSUE BASED ON MEAN SOLUBLE MERCURY FOR SITES 2,3,4, AND 6.

calibrated model predicted the mercury in filet values for fish at sites 3, 4, and 6 within the confidence limits of the data, the model was accepted as verified.

Verification of Zero Order Kinetics for Model Constants

The equilibrium model employed assumes that the rate of absorption at any particular water concentration is a constant and not affected by the concentration reached by the organism. That is, the uptake rate (kw) is proportional only to the concentration of mercury in the water. The existence of zero order kinetics can be tested by rearranging equation (5) as:

$$C = k_w C_w [1 - \exp(-k_d t)]/k_d$$

Table 5 summarizes the calculations for sites 2, 3, 4, and 6. A plot of the function in brackets against C should yield a linear relationship if both k_w and C_w are constant. Figure 8 demonstrates such a plot for each of the study sites. Note that each of the four sites give a straight line relationship, therefore, zero order kinetics is verified and the use of the simple steady state kinetic model employing soluble water column mercury to predict mercury accumulation for the system is justified.

Model Predictions of Methylmercury Concentrations in EFPC

The water column methylmercury data developed during this study was very limited, i.e., two data points for site 2; single data points for sites 1, 3, 4, 5, and 6, and control. All samples were collected at the end of the study period in December. Because water column methylmercury

TABLE 5 SUMMARY OF DATA FOR VERIFICATION OF ZERO ORDER KINETICS FOR CONSTANTS ${\bf k_W}$ AND ${\bf k_d}$ (SOLUBLE MERCURY MODEL)

		Average	Hg in Muscle Tiss	ue by Site (ug/g)	
!	l-e ^{-k} d ^t	Site 2	Site 3	Site 4	Site 6
t	k _d	$(C_W = 0.32 \text{ ug/1})$	$(C_W = 0.13 \text{ ug/l})$	(C _W = .082 ug/1)	(C _W = .06 ug/l)
22	19.7	.087	.037	.027	.023
44	35.6	.198	.075	.039	.034
64	47.3	.305	.114	.078	.044
86	57.7	.480		.132	.046
108	66.0	.488			.056
136	74.3	.518	; 		

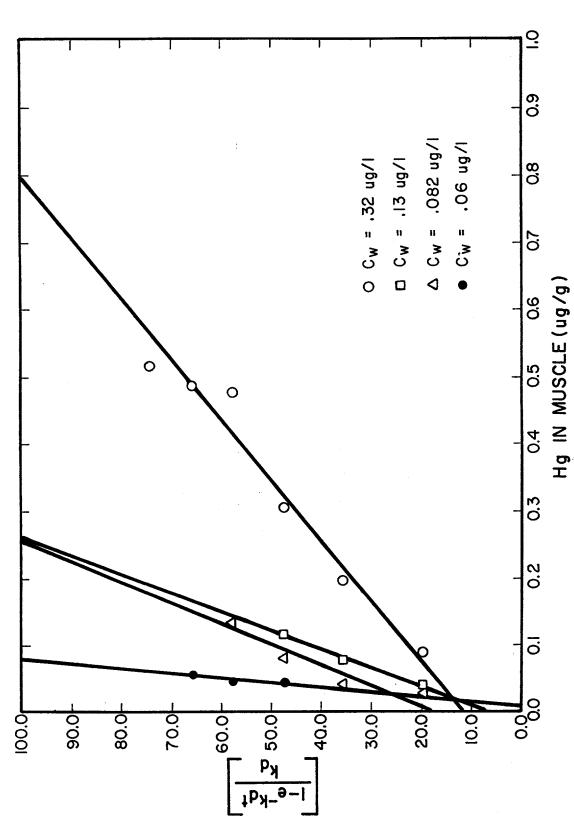


FIGURE 8 DEMONSTRATION OF ZERO ORDER KINETICS FOR THE UPTAKE CONSTANT kw FOR SITES 2,3,4, AND 6

concentrations are of primary concern when modeling mercury accumulation by fish, an estimate of dissolved methylmercury is necessary to assess the EFPC contamination problem. One way to estimate dissolved methylmercury is to select literature values for the model coefficients k_d and k_w , and knowing the mercury concentration in fish muscle tissue at a specific time, calculate the water column concentration for methylmercury. For example, Hartung (1976), provides estimates of depuration of methylmercury to be 0.00099 d^{-1} and a bioconcentration factor of 1,650 at 20° C). Therefore:

$$C_t = C_w BCF (1-e-k_d t)$$

Or:

$$C_w = C_t / BCF(1-e-k_dt)$$

Employing the average muscle tissue mercury values at Day 64 for sites 2, 3, 4, and 6 (see table 5) and the literature values for methylmercury depuration and bioconcentration, the above equation was used to estimate the soluble methylmercury exposure at each site. The estimated values were 30 ng/1, 11.2 ng/1, 7.7 ng/1 and 4.3 ng/1 for sites 2, 3, and 4, respectively. The estimated values are 2 to 4 orders of magnitude higher than those measured at these sites. Hartung (1976) applied this technique to Cayuga Lake trout data and estimated the average methylmercury water column concentration to be 7.3 ng/1, which gives some support to these numbers. Reducing the values of the model constants, to account for the 2 to 4 order of magnitude difference, is not justified because the values used are near the limits reported in the literature. The differences between observed and predicted values could be due to

analytical problems for methylmercury and the fact that the uptake from food sources are not known.

Contribution of Mercury From Food

The amount of mercury contributed by food intake may be estimated by:

$$\frac{AE * R * C_{f}}{k_{d}} (1 - e^{-k} d^{t})$$
 (8)

Where:

AE = assimilation efficiency (%)

R = food ration (g/g-d)

 C_f = mercury concentration in ration (ug/g)

Usually AE is determined experimentally by comparing the steady-state body burdens in "water-only" and "food-plus-water" treatments. However, because these data sets were not available, and 81 percent of the fish in the cages sampled showed significant weight loss (10 to 20 percent was common), mercury uptake from food was considered an insignificant source.

However, an estimate of the contribution from food for natural fish populations can be made using the following model assumptions, i.e., daily food ration equals 3 percent or 0.03 g/g * d and $C_{\hat{f}}$ equals 6.6 ug/g total mercury (average of invertebrate data from Appendix A) and an assimilation efficiency of 80 percent. The estimate of the constant,

 k_d , (0.01 d⁻¹) was calculated as previously described. Substituting these values in Equation 8 for t = 136 days, yields:

$$\frac{AE * R * C_f}{k_d} (1-e^{-k}d^t) = 2.9 \ \mu g/g \qquad (9)$$

The estimated potential contribution from food after 136 days, 2.9 ug/g, is an order of magnitude higher than the measured value for the site 2 data (i.e., the site showing the highest values). Although it is not possible to determine what portion of the measured filet concentration came from water or from food, the analysis does support the previous observation that they were not actively feeding in the cages. The analysis also suggests that food could be a significant factor for mercury accumulation of the natural fish populations of EFPC.

Model Assumptions and Limitations

The following assumptions and limitations must be considered when evaluating the model results.

- 1. The model is steady-state and therefore does not represent the dynamic flux of water column mercury or the feeding habits of fish. However, the water column data for most sites did not show variations of the extreme, which would invalidate the steady-state approach.
- 2. The model parameters are valid for the age of test fish (unknown). The assumption that bluegill/green sunfish hybrid will exhibit the most dramatic response in mercury accumulation in EFPC may not be valid and, therefore, the model should not be interpreted between species.

- 3. Growth Rate: The model does not account for the dilution effect of fish growth at this time. By excluding the growth rate we are examining the worst case scenario.
- 4. Fixed Depuration Rate and Assimilative Efficiency: The chosen depuration rate and assimilative efficiency are estimated values. Depuration rate may vary with temperature and fish age, and the mode of uptake, although the literature is ambiguous on these points.
- 5. The model uses soluble water column mercury to predict mercury uptake by fish, which suggests soluble mercury can be used as a surrogate for the more difficult to measure methylmercury.

SUMMARY

The following conclusions are based on review of the current literature, results of research projects conducted by ORNL, and the modeling results described in this report.

- Total water column mercury concentrations are elevated at each of the EFPC sites studied.
- The source of mercury in the EFPC system is apparently from past and ongoing mercury discharge from the Y-12 plant.
- 3. The primary source of mercury uptake by bluegill is total mercury in the water column of EFPC.

- ciated with suspended sediments composed of colloidal clay and silt fractions that due to their common charge resist settling even in ponded areas and, consequently, are continually associated with the water column. Mercury associated with the suspended sediments may exchange directly to the fish when these contaminated particles are filtered by the filaments of the gills. Gills are the site of gas exchange (respiration) for fish and provide a large surface area which is separated from the water and particles by a single cell membrane. Evidently, mercury is exchanged from the sediment particles to (or through) the cell membrane to the fish blood. The efficiency of this flux is undetermined.
- The sediment-water flux rate, i.e., the amount of total mercury and methylmercury released per unit area of sediment per unit column of water per day, probably is an important mechanism affecting mercury uptake by fish, and therefore, is an important data need.
- 6. Site 1 total water column mercury concentrations averaged
 3.6 ug/1 during the study period and EFPC flow averaged
 27 cfs. Because site 1 is located above New Hope Pond and
 flow in EFPC at this point is dominated by Y-12 discharge
 (up to 20 cfs), the estimated mercury release from the plant
 during the study period was approximately 0.4 lb/day.

RECOMMENDATIONS

Considering the level of mercury contamination in upper EFPC from previous discharges and the continued discharge of mercury at levels which appear to be above those of current treatment technology, it is unlikely that FDA limits for mercury in fish filets will be met without remedial measures.

The following studies are recommended:

- 1. Studies are needed to identify areas of significant mercury methylation, the relationships of mercury methylation, total and soluble water column mercury, and the uptake of mercury by fish. Can soluble water column mercury be used as a surrogate parameter to estimate methylmercury uptake as suggested by the model in this report?
- Definitive studies are needed to determine the significance of mercury uptake from fish food sources.
- 3. Descriptive studies of mercury levels in other species of fish in EFPC (i.e., concentration by age class and by location) to verify that bluegill are the most sensitive species, to monitor mercury accumulation in fish in EFPC.
- 4. PCB and other contamination in the system should be evaluated in the same manner, in order that remedial measures reflect all significant contaminants.

FAEB 0410h

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APPENDIX A

MERCURY CONCENTRATIONS IN FILLETS FROM BLUEGILL AND GREEN SUNFISH HYBRIDS FROM EAST FORK POPULAR CREEK - 1986

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Avg Tot H (ug/g)			၀ က ျ	.023	. 11.	0.198
CF	1 4 1 1 1 1	1.054 1.054 1.059 1.000 1.017	1.132 1.234 1.135 1.047 0.986	1.015 0.958 0.958 0.958 0.958 0.928 0.929 0.929	11.1.1.1.1.1.1.1.1.1.1.1.1.1.1.1.1.1.1	1.065 1.179 1.039 0.955 0.913
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TOTAL HG RATE (ug/g/d)					0.00398 0.00398 0.00398 0.00382 0.00382 0.00164 0.00223	
TOTAL HG (ug/g) (0.008 0.015 0.015 0.014 0.011	0.051 0.053 0.053 0.073	0.003 0.032 0.033 0.035	0.020 0.027 0.023 0.023 0.015 0.016 0.028	0.123 0.195 0.195 0.112 0.221 0.183 0.112	0.250 0.250 0.219 0.073
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CF				0.920 0.920 0.920 0.920 0.923 0.923	
BODY BURDEN (49)				10.00 10.00	
TOTAL HG RATE (ug/g/d)		0.00379 0.00558 0.00589 0.00158 0.00123 0.00123	0.00030 0.00042 0.00030 0.00123 0.00123 0.00228 0.00333	0.00013 0.00013 0.00013 0.00013 0.00013	000000000000000000000000000000000000000
TOTAL HG (ug/g)	0000000	0.330 0.150 0.150 0.120 0.120 0.140	oooo a dhi u n	400000	00000000000000000000000000000000000000
CHRNGE LENGTH (CR)	0 6 4 4 6 0 4 6 6 6 6 6 6 6 6 6 6 6 6 6 6 6 6 6 6 6		0000000		
FINEL LENGTH					
INITAL LENGTH (CH)	22.22.22.22.22.22.22.22.22.22.22.22.22.	V	0.0.0.0.0.0.0.0.0.0.0.0.0.0.0.0.0.0.0.		
CHRNGE NT (9)	1 W 1 W 1 W 4 W 4 W 4 W 4 W 4 W 4 W 4 W	04410016 65			
FINAL HT (9)	2 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4	4 W 4 W 4 W 9 W 9 W 9 W 9 W 9 W 9 W 9 W	0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	. 400 00 00 00 00 00 00 00 00 00 00 00 00	
INITAL HT (9)	25.25 20.25	m d d d d d d d d d d d d d d d d d d d	200 4 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2	4 6 6 4 6 6 4 6 6 6 6 6 6 6 6 6 6 6 6 6	- 4
TINE	6 8 8 8 8 8 8 8 8 8 8 8 8 8 8 8 8 8 8 8	### ### ##############################	888888555		98 8 8 8 8 8 8 8 8 8 8 8 8 8 8 8 8 8 8
DATE	1 - 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4	0.000000000000000000000000000000000000	22 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2		11-11-13-14-14-14-14-14-14-14-14-14-14-14-14-14-
CAGE?	<u> </u>	7 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7	ក្នុង	រត់សិសសិសសិសសិសសិស	<u> </u>
SITE	9	<i>.</i>	<i>റ</i> ഗഗതനയയയ		144444444444444444444444444444444444444
TAG #	622 15 15 16 106 117	179 192 211 211 364 343 343 NT-2 391 556	625 629 612 655 299 299 299 127 1127	NT-1824 610 610 610 633 653 641 658 658 658	200 191 191 191 193 193 193 193 193 193 193

	ñ																					
1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1	. B00Y	BURDEN (40)														:		:				
	TOTAL	Cug/q/d)		-0.00014	-0.00014	-0.00014	-0.00014	-0.00014	-0.00014	ERR	ERR	ERR	ERR	ERR	ERR	ERR	ERR	ERR	ERR	ERR	ERR	
	TOTAL	6/67 5									6. 2.	7	ນຸ	6.2	31	1.8	0.61	2.9	4.2			
	CHANGE	(CH)	1												,	***						
1,111111	FINGE	((0))		r .	13.6	F. F.		13.8	5													
	INITAL	, CHU)	1. 6	11.0	0 6	14.3	¥ C	7.00	16.1													
11111111111	CHANGE	6																				
	FINAL	6	4.04	50	, C	7 V	. Δ . α	. K.	}							ı						
	INITAL HT	Ç	36.3	4	42.7	39.6	39.1	13. E	2													
1	CDRYS		101	101	101	101	101	101	! !													
	OUT	1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1	10-9-86	10-9-86	10-9-86	10-9-86	10-9-86	10-9-86		9-12-86	9-12-86	9-12-86	9-12-86	9-12-86	9-12-86	9-12-86	9-12-86	9-12-86				
STTE COCES	ב ממני	•	오	웆	2	웃	웆	웆														
STTE	1	٥	N	N	N	~	ď	N			~			-	N	~	N	N				
	TAG #	1111111	&	13	ee	543 543	476	250		Lgdrfl	Ldrgfl	Danslfl	Dams1f1	Snails	Lgdrf1	Dans1f1	Crayfish	Snails				

SITE	DATE	TEMP	COND (uS/cm)	рН	D8 (mg/L)	TOT HG	DIS HG
			· ····································		(mg/L)	(ug/L)	(ug/L)
1	08-Ju1-86	25.4	402	8.06	7.8	2.6	1.3
2	08-Ju1-86	25.6	378	8.75	8.3	1.4	0.3
3	08-Ju1-86	24.1	379	8.32	8.1	1.4	0.3
4	08-Jul-86	22.9	413	8.13	7.9	1.2	0.07
5	08-Ju1-86	23.9	409	8.08	7.6	1.1	0.05
ሪ	08-Ju1 -8 6	22.5	432	7.92	6.8	0.7	0.04
· 1	30-Jul-86	23.8	375	8.35	6.9	3.7	0.04
2	30-Jul-86	22.9	346	8.59	6.4	1.8	0.5
3	30-Jul-86	22.1	-348	8.08	6.4	1	0.8
4	30-Ju1-86	21.8	363	8.16	9.7	0.6	0.1
5	30-Jul-86	22.3	418	7.93	9.1	1	0.05
· 1	21-Aug-86	26.6	.338	8.23	8.2	2.74	0.58
· 2	21-Aug-86	26	374	8.88	10.8	1.3	0.35 0.37
3	21-Aug-86	24.4	389	8.14	8.3	0.81	0.12
4	21-Aug-86	23.9	361	8.14	7.4	0.56	0.05
6	21-Aug-86	23.4	387	7.73	5.4	0.68	0.02
1	10-Sep-86	24.5	379	8.17	7.7	7.5	6.1
2	10-Sep-86	22.8	347	8.74	10.45	1.3	0.31
3	10-Sep-86	21.7	374	8.15	8.45	0.8	0.15
4	10-Sep-86	21.3	348	8.15	7.9	0.56	0.11
· 6	10-Sep-86	20.3	400	7.67	5.6	0.53	0.14
1	26-9ep-86	26.3	1065	9.76	5.8	2.8	0.18
2	26-Sep-86	25.2	430	8.45	8.9	1.4	0.19
3	26-9ep-86	23.8	419	8.21	7.7	1.5	0.11
4	26-Sep-86	23.5	1 41	8.15	7.2	0.49	0.08
6	26-Sep-86	22.5	475	7.94	6.7	0.43	0.05
1.	16-Oct-86	21	416	7.94	6.6	2.82	0.59
2	16-0ct-86°	18.3	443	8.11	8.4	1.25	0.28
3	15-Oct-85	16.5	453	8.1	9.3	0.99	0.16
4	16-0ct-86	13.8	477	8.01	8.2	0.63	0.09
6	16-Oct-86	14.3	510	7.9	3.6	0.51	0.04
1	14-Nov-86	18.8	492	7.92	7.9	1.8	0.26
2	14-No∳-86	16.2	521	7.85	8	1.3	0.09
1	10-De ¢ -86	16.7	417	7.83	8	5	ំ.ន
2	10-Dec-86	15.3	379	7.79	8.6	2.3	0.5
3	12-Dec-86	10.2	297	7.74		0.92	0.09
4	12-Dec-36	10.4	392	7.73	7.4	0.94	0.05
	12 ec-85	11-3	141	7.74	9	0.02	0.01
5	15-pec-86	8.3	415	7.91	9.1		0.04
6	15-Dec-84	9.8	405	7.84	8.8	0.29	0.04
2	17-Dec-86	14.8	451	7.73	3.7	- w ,	= - ·- ·

			•			
÷	SITE	DATE	TEMP	TOT HG	DIS HG	Me-Hg
		•	(C)	(ug/L)	(ug/L)	(ng/L)
				3		g,
	1	08-Jul-86	25.4	2.6	1.3	-
	2	08-Ju1-86	25.6	1.4	0.3	
	3	08-Jul-86	24.1	1.4	0.2	
	4	09-Ju1-86	22.9	1.2	0.07	
	5	08-Ju1-86	23.9	1.1	0.05	
	6	08-Ju1-86	22.5	0.7	0.04	
	· 1	30-Ju1-86	23.8	3.7	0.9	
	2	30-Ju1-86	22.9	1.8	0.6	***
	3	30-Ju1-86	22.1	. 1	0.2	
	4	30-Ju1-86	21.8	.0.6	0.1	
	6	30-Jul-86	22.3	1	0.05	
	1	21-Aug-86	26.6	2.76	0.58	
٠	2	21-Aug-86	26	1.3	0.37	***
	3	21-Aug-85	24.4	0.81	0.12	-
	. 4	21-Aug-86	23.9	0.56	0.05	*
	6	21-Aug-86	23.4	0.68	0.02	
	1	10-Sep-86	24.5	7.5	6.1	-
	2	10-Sep-84	22.8	1.3	0.31	
•	3	10-Sep-86	21.7	0.8	0.15	
	4	10-Sep-86	21.3	0.56	0.11	****
-	6	10-Sep-86	20.3	0.53	0.14	grandy warms
#	1 -	26-Sep-86	26.3	2.8	0.18	
	2	26-Sep-86	25.2 `	1.4	0.19	
	3	25-Sep-85	23.8	1.5	0.11	
	4	26-Sep-86	23.5	0.49	0.08	
	5	26-Sep-86	22.5	0.43	0.06	
	1	16-0ct-86	21	2.92	0.59	
	3 3	16-Oct-86	18.3	1.25	0.28	
	3 4	16-Oct-86	16.5	0.99	0.16	
	4 5	16-0ct-86	13.8	0.63	0.09	
	1	16-0ct-86	14.3	0.51	0.04	
	5	14-Nov-86 14-Nov-86	18.8	1.8	0.26	
	1	10-De é -86	16.2	1.3	0.09	
	5	10-De ć -85	15.7	. 5	o.s	0.2
	3	12-Dec-86	15.3 10.2 °	2.3	0.5	0.5
	4	12-Dec-85		0.93	0.09	0.4*
C	Control	12-Dec-85	10.4 11.3	0.94	0.05	ND
_	. 5	15-Dec-86	8.3	0.02	0.01	NE
	5	15-De c -85	9.8	0.37	0.04	0.4*
	2	17-Dec-86	14.8	0.29	0.04	0.2
_	-	r, per og	1.4.0	૩. દર	0.45	0.8

^{*} Samples analyzed in duplicate; one of the samples was below the detection limit, the other had the value given in the table.